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Renormalization of the tunnel splitting in a rotating nanomagnet

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We study spin tunneling in a magnetic nanoparticle with biaxial anisotropy that is free to rotate about its anisotropy axis. Equations of motion are derived that couple spin and mechanical degrees of freedom and exact instanton of these equations is obtained. We show that mechanical freedom of the particle renormalizes magnetic anisotropy and increases the tunnel splitting.

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Macroscopic dynamics of a fixed-length magnetic moment, \( \mathbf{M} \), of a single-domain ferromagnetic particle is described by the Landau-Lifshitz equation\(^3,4\). When dissipation (which is usually weak) is neglected this equation reads

\[
\frac{\partial \mathbf{M}}{\partial t} = \gamma \mathbf{M} \times \mathbf{B}_{\text{eff}}, \quad \mathbf{B}_{\text{eff}} = -\frac{\delta \mathcal{E}}{\delta \mathbf{M}}, \tag{1}
\]

where \( \mathcal{E} \) is the classical magnetic energy of the particle that depends on the orientation of \( \mathbf{M} \). It was shown long ago\(^3,4\) that Eq. (1), besides the real-time solutions, also possesses imaginary-time solutions - instantons - that describe macroscopic quantum tunneling of \( \mathbf{M} \) between classically degenerate energy minima (see also books: Refs. 2 and 5). They provide the tunnel splitting of classically degenerate spin states. Magnetic instantons have been intensively studied for various symmetries of the crystal field Hamiltonian and also for problems involving spatial dependence of the spin field, such as, e.g., tunneling of domain walls and nucleation of magnetic bubbles\(^5\). The effect of interaction of the tunneling spin with the dissipative environment (phonons, nuclear spins, etc.) has been another active field of study.

In early experiments on spin tunneling\(^5\), single-domain magnetic particles were always frozen in a solid matrix so that their physical position and orientation were fixed and only rotation of the magnetic moment was allowed. Later, beams of small magnetic clusters were investigated\(^7–12\) and more recently free magnetic nanoparticles confined within solid nanocavities have been studied\(^6\). Experimentalists have also worked with molecular nanomagnets deposited on surfaces\(^13–16\) or carbon nanotubes\(^17\), as well as with single magnetic molecules bridged between metallic electrodes\(^18–22\). In such experiments the particles retain some mechanical freedom. This inspired recent theoretical work on quantum mechanics of rotating magnets\(^23–26\). In this paper we study the tunnel splitting in a nanomagnet that is free to rotate about its magnetic anisotropy axis. An interesting example of exact magnetic instanton will be given that mixes the spin and mechanical degrees of freedom. Our results may be relevant to recent experiments with single magnetic molecules.

As is well known, even the classical problem of the rotation of a rigid body is rather involved. In a quantum problem the energy levels related to the rotational degrees of freedom can be obtained analytically only for a symmetric body\(^27\). Even this simplified problem becomes significantly more complicated when the rotating body possesses a spin. Nevertheless, as was recently demonstrated in Ref. 23, the exact low-energy eigenstates can be obtained analytically for a nanomagnet that, due to a large magnetic anisotropy, can be described as a two-state spin system and is free to rotate about its magnetic anisotropy axis. It was shown that the crucial role is played by the parameter

\[
\alpha = 2(hS)^2/(I\Delta). \tag{2}
\]

Here \( S \) is the dimensionless total spin of the nanomagnet, \( I \) is its moment of inertia, and \( \Delta \) is the spin-tunnel splitting. At \( \alpha < \alpha_1 \equiv \left[ 1 - 1/(2S)^2 \right]^{-1} \) the ground state and the first excited state are respectively symmetric and antisymmetric superpositions of \( J = L + S = 0 \) states shown in Fig. 1. Such superpositions are characterized by zero magnetic moment. At \( \alpha > \alpha_1 \) the nanomagnet develops a finite magnetic moment. In the limit of \( \alpha \rightarrow \infty \) the spin...
localizes in one of the two directions along the magnetic anisotropy axis\textsuperscript{23}. This can be understood qualitatively based upon the following argument. Delocalization of \textbf{S} in the spin space due to tunneling transitions between \(|S\rangle\) and \(|-S\rangle\) lowers the ground state energy by \(\Delta/2\). However, for a magnet that is free to rotate this can only occur if the spin tunneling is accompanied by cotunneling of \textbf{L}, so that \(\textbf{J} = \textbf{L} + \textbf{S}\) is conserved, Fig. 1. If the corresponding mechanical energy, \((\hbar S)^2/(2I)\), is large compared to \(\Delta/2\), which is the case of large \(\alpha\), then tunneling cannot lower the energy and the spin localizes in one of the \(|\pm S\rangle\) states.

In this paper we ask the question that has not been previously addressed. How does the mechanical freedom of the nanomagnet change the tunnel splitting \(\Delta\)? When rotations are treated semiclassically the answer to this questions can be obtained by studying instantons that connect the two states shown in Fig. 1. This problem is rather interesting as the corresponding instantons should mix spin and mechanical degrees of freedom. It also has practical implications for experimental studies of magnetic molecules and atomic clusters that are free to rotate. Such molecules or clusters should have very weak anisotropy that is free to rotate about its easy axis \(X\)-axis. This can be, e.g., a small atomic cluster held inside an asymmetric trap or a nanoparticle. Anisotropy constants \(K_{\perp} > 0\) and \(K_{\|} > 0\) have been redefined to show the explicit proportionality of the anisotropy energy to the volume. They are dimensionless numbers, typically of order unity.

The rotational kinetic energy of the particle is \(E_R = \frac{1}{2}I\dot{\phi}^2\). The Lagrangian of our system consists of the trivial kinetic Lagrangian for the variable \(\varphi\),

\[
L_L = \hbar \textbf{L} \cdot \dot{\varphi} - E_R = \frac{1}{2}I\dot{\varphi}^2, \quad (4)
\]

and the magnetic Lagrangian,

\[
L_S = \left(\frac{M_0V}{\gamma}\right)\dot{\varphi} \cos \theta - E_A(\theta, \phi), \quad (5)
\]

for the variables \(\theta\) and \(\phi\). The first term in Eq. (5) follows from the fact that \(\hbar S_z = \hbar S \cos \theta\) is the generalized momentum for the coordinate \(\phi\). The second term is the effective magnetic energy in the rotating frame,

\[
E_A = E_A(\theta, \phi) - \hbar \textbf{S} \cdot \dot{\varphi}. \quad (6)
\]

The last term in this equation is related to the fact that in the particle frame the rotation of the magnetic field \(\textbf{B} = \dot{\varphi}/\gamma\). The total Lagrangian is a sum of \(L_L\) and \(L_S\):

\[
L = \hbar (L+S) \cdot \dot{\varphi} + \frac{M_0V}{\gamma} \dot{\varphi} \cos \theta - \left[\frac{1}{2}I\dot{\varphi}^2 + E_A(\phi, \theta)\right]. \quad (7)
\]

The first term reflects the fact that in the presence of a spin the generator of rotations is \(\textbf{J} = \textbf{L} + \textbf{S}\). The explicit form of the total Lagrangian in terms of the generalized coordinates \(\theta, \phi, \text{ and } \varphi\) is

\[
L = \frac{1}{2}I\dot{\varphi}^2 + \left(\frac{M_0V}{\gamma}\right)\dot{\varphi} \cos \theta + \left(\frac{M_0V}{\gamma}\right)\dot{\varphi} \sin \theta \cos \phi
\]

\[
- \frac{1}{2}\mu_0M_0^2V[(K_{\perp} + K_{\|} \cos^2 \phi) \cos^2 \theta + K_{\|} \sin^2 \phi]. \quad (8)
\]

The equations of motion are Euler-Lagrange equations for \(\theta, \phi, \text{ and } \varphi\):

\[
\frac{d\phi}{dt} = (K_{\perp} + K_{\|} \cos^2 \phi) \cos \theta + \dot{\phi} \cos \theta \cos \phi \sin \theta \quad (9)
\]

\[
\frac{d(\cos \theta)}{dt} = -K_{\|} \cos \phi \sin \phi \sin^2 \theta - \dot{\phi} \sin \theta \sin \phi \quad (10)
\]

\[
\frac{d}{dt} \left[ I\dot{\varphi} + \frac{V}{\mu_0\gamma^2} \cos \phi \sin \theta \right] = 0. \quad (11)
\]
where we have introduced dimensionless time $\tilde{t} = \gamma \mu_0 M_0 t$ and $\tilde{\phi} = d\phi/d\tilde{t}$.

Note that the equations of motion for $\phi$ and $\theta$ can also be obtained from the Landau-Lifshitz equation with $\mathcal{E} = E_A'$. Indeed, the equations for $\phi$ and $\theta$ that follow from Eq. (1) (see, e.g., Ref. 2),

$$\frac{\partial \phi}{\partial t} = -\frac{\gamma}{M \sin \theta} \left( \frac{\partial E_A'}{\partial \phi} \right), \quad \frac{\partial \theta}{\partial t} = \frac{\gamma}{M \sin \theta} \left( \frac{\partial E_A'}{\partial \phi} \right),$$

are identical to the equations (9) and (10). The third equation of motion, Eq. (11), is the conservation of the total angular momentum: $\frac{d}{dt}[L_X + S_X] = \frac{d}{dt}J_X = 0$. At $J_X = 0$ it is equivalent to the constraint:

$$I \dot{\phi} = -\frac{M_0 V}{\gamma} \sin \theta \cos \phi.$$

With account of this constraint the equations of motion for $\phi$ and $\theta$ become

$$\frac{d\phi}{dt} = (K_\perp + K'_\parallel \cos^2 \phi) \cos \theta,$$

$$\frac{d(\cos \theta)}{dt} = -K'_\parallel \cos \phi \sin \phi \sin^2 \theta,$$

where

$$K'_\parallel = K_\perp - K_R, \quad K_R = \frac{V}{\mu_0 \gamma^2 I}.$$

We see that for $J = 0$ the effect of rotations reduces to the renormalization of the easy axis anisotropy $K_\parallel$. This is easy to understand from the following consideration. In a state with $J_x = 0$, equilibrium vectors $S$ and $L$ look in the opposite directions along the $x$-axis. If $S$ deviates from the $x$-axis, $S_x$ decreases and so should $L_\perp$ to preserve the condition $J_x = 0$. The decrease of $L_\perp$ corresponds to the decrease of the rotational energy, $(hL_x)^2/(2I)$, mandated by $J_x = 0$. Thus, effectively, the magnetic anisotropy energy associated with the deviation of $S$ from the easy axis becomes smaller when mechanical rotation is allowed.

We should now look for solutions of equations (14) and (15). We first notice that

$$\mathcal{E} = \frac{1}{2} \mu_0 M_0^2 V [(K_\perp + K'_\parallel \cos^2 \phi) \cos^2 \theta + K'_\parallel \sin^2 \phi]$$

is the integral of motion. This is easy to see by differentiating this equation on time and substituting in the resulting equation the time derivatives $\phi$ and $\theta$ from equations (14) and (15). Not surprisingly, up to a constant, Eq. (17) equals the total energy of the particle in the laboratory frame, $\mathcal{E} = \mathcal{E}_A + \frac{1}{2} I \dot{\phi}^2$, with account of the constraint (13). Eq. (14) gives

$$\cos \theta = \frac{d\phi/d\tilde{t}}{K_\perp + K'_\parallel \cos^2 \phi}.$$

This allows one to express $\mathcal{E}$ in terms of the angle $\phi$ and its time derivative:

$$\mathcal{E} = \frac{1}{2} M_0^2 V \left[ (d\phi/d\tilde{t})^2 + K'_\parallel \cos^2 \phi + K'_\parallel \sin^2 \phi \right].$$

Since this expression is positively defined (see discussion after Eq. (26)), the classical energy minima occur at $\mathcal{E} = 0$. They correspond to the stationary magnetization pointing in either direction along the easy axis, i.e., $\phi = 0, \pi$ with $\cos \theta = 0$ in accordance with Eq. (18).

Equation $\mathcal{E} = 0$ has no real-time solutions for $\phi$ that connect the two degenerate classical energy minima. However, in imaginary time, $\tilde{t} = i\tilde{t}$, equation $\mathcal{E} = 0$ is equivalent to

$$\left( \frac{d\phi}{d\tilde{t}} \right)^2 = K'_\parallel \sin^2 \phi (K_\perp + K'_\parallel \cos^2 \phi).$$

Such equation has instanton solutions that connect the classical energy minima:

$$\phi(\tau) = \pm \arccos \left( -\frac{\sinh(\omega_0 \tau)}{\sqrt{\lambda + \cosh^2(\omega_0 \tau)}} \right).$$

The $\tau$-dependence of $\theta$ is given by Eq. (18), and the $\tau$-dependence of $\phi$ is given by Eq. (13). Here

$$\lambda = K'_\parallel/K_\perp, \quad \omega_0 = |\gamma| \mu_0 M_0 \sqrt{K'_\parallel (K'_\parallel + K_\perp)}.$$

Positive and negative signs correspond to the two possible trajectories, which are counterclockwise and clockwise rotations of the magnetization from $\phi = 0$ at $\tau = -\infty$ to $\phi = \pm \pi$ at $\tau = +\infty$, respectively, see Fig. 2.

The tunnel splitting has the form $\Delta = A e^{B}$, where $A$ is of the order of quantized oscillations near the minimum of the potential well and $B = \oint_{-\infty}^{+\infty} d\mathcal{E}$ is the WKB exponent. Substituting here $\mathcal{E}$ of Eq. (7) at $J = 0$, one obtains for the instanton trajectory

$$B = -S \ln \left( \frac{\sqrt{1+\lambda} + \sqrt{\lambda}}{\sqrt{1-\lambda}} \right).$$
In a microscopic theory the easy-axis crystal field is presented as $-DS^2$. The connection between $D$ and the parameter $K_\parallel$ of the macroscopic theory is

$$K_\parallel = \left(2 - \frac{1}{s}\right) \frac{D V_0}{\mu_0(h \gamma)^2}, \quad (24)$$

where $s$ is spin per unit cell of the crystal and $V_0$ is the volume of the unit cell. (Singularity at $s = 1/2$ reflects the fact that single-ion magnetic anisotropy does not exist for spin $1/2$). The total spin of a ferromagnetic particle can be presented as $S = s(V/V_0)$. Consequently,

$$\frac{K_R}{K_\parallel} = \frac{S h^2}{(2s - 1) J D} = \frac{\Delta}{(2s - 1) 2SD} \alpha, \quad (25)$$

where we have used Eqs. (2) and (16). Renormalization of the easy-axis anisotropy by rotations can be presented in the form $K'_\parallel = K_\parallel (1 - \epsilon)$, where

$$\epsilon = \left(\frac{\alpha}{2s}\right) \left(1 - \frac{1}{2s}\right) \frac{\Delta}{E_1} \quad (26)$$

and $E_1 = (2s - 1)D$ is the energy of the first excited spin state at $\Delta = 0$.

The low energy limit that we are studying corresponds to $\Delta \ll E_1$ and $\alpha < \alpha_1$. In this limit $\epsilon$ is small. Consider, e.g., the case of large $S$ and large $\lambda$ (small tunneling rate). According to Eq. (23) in this case $B = -S \ln(4\lambda)$ so that $\Delta \propto \exp[-S \ln(4\lambda)]$. It is easy to see from this expression that mechanical rotation renormalizes $\Delta$ by a factor $\exp(\epsilon S)$. Normally it would not be large compared to one. However, since small $\epsilon$ in the exponent is multiplied by a large $S$, it is not out of question that at sufficiently large $\Delta$ a slight increase of the tunnel splitting would be observable in ESR studies of spin clusters that are free to rotate. For, e.g., a typical magnetic molecule with $S = 10$, $I \sim 10^{-35} \text{ g \ cm}^2$, and $E_1 \sim 10\text{K}$, the condition $\alpha = \alpha_1$ corresponds to $\Delta \sim 0.1\text{K}$ and $\epsilon S \sim 0.1$, which would result in small but experimentally detectable difference in the tunnel splitting for the molecule firmly coupled to a solid and the molecule that is free to rotate.

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